A review on membrane fouling control in anaerobic membrane bioreactors by adding performance enhancers

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Abstract

Anaerobic membrane bioreactors (AnMBRs), the combination of anaerobic digestion and membrane technology, have gained increasing popularity due to their remarkable advantages over aerobic membrane bioreactors, such as biogas production and potential energy use. However, membrane fouling remains a challenging issue that deteriorates the performance of membrane and shortens its lifespan. Pretreatment of feed wastewater by adding fouling reduction enhancers, such as adsorbents and flocculants, into anaerobic membrane bioreactor

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Abbreviations: AnMBR, anaerobic membrane bioreactor; PAC, powder activated carbon; GAC, granular activated carbon; SRT, solids retention time; VFAs, volatile fatty acids; MBRs, membrane bioreactors; AC, activated carbon; DOM, dissolved organic matters; BAC, biologically activated carbon; EPS, extracellular polymeric substance; DMBR, aerobic dynamic membrane bioreactor; TMP, transmembrane pressure; COD, chemical oxygen demand; SMP, soluble microbial products; SMX, sulfamethoxazole; TC, tetracycline; ETS, erythromycin-tetracycline-sulfamethoxazole; ST, erythromycin-tetracycline; Tmp, trimethoprim; Cbz, carbamazepine; Dcf, diclofenac; Tcs, triclosan; DOC, dissolved organic carbon; EGSB, expanded membrane-coupled granular sludge bed; DIET, direct interspecies electron transfer; UASB, upflow anaerobic sludge blanket; LCWW, low-grade coal wastewater; ZVI, zero-valent iron; ORP, oxidation-reduction potential; MLSS, mixed liquor suspended solids; TP, total phosphorous; EGSB, expanded granular sludge bed; SCFA, short chain fatty acids; LCFA, long chain fatty acids; SS, suspended solid; AOX, adsorbable organic halogen

can effectively mitigate membrane fouling by altering the feed properties. Activated carbon, such as powdered activated carbon (PAC) and granular activated carbon (GAC), has been widely applied as an adsorbent to aerobic and anaerobic membrane bioreactors for membrane fouling control. Organic enhancers such as biochar and waste yeast, and inorganic enhancers like polyaluminum chloride and zeolite have also been applied to AnMBRs promoting flocculation and coagulation. Thus, this review discusses the impacts of different fouling reduction enhancers under anaerobic conditions as well as AnMBR system. In addition, the mechanisms of the enhancers mitigating the membrane fouling are also summarized for better understanding of the effects of the enhancers in AnMBRs.

Keywords: Anaerobic membrane bioreactor; membrane fouling; enhancers; activated carbon

1. Introduction

In recent years, anaerobic membrane bioreactor (AnMBR) technology, which combines anaerobic process and membrane filtration, has been gaining increasing popularity. AnMBRs have the same benefits as aerobic MBRs, such as a footprint reduction and superior permeate quality. AnMBRs also can provide several advantages over the aerobic processes, including long solids retention time (SRT), low sludge production and potential energy use [1]. Moreover, as AnMBR is the integration of anaerobic digestion process with membrane separation, these processes can provide benefits as well. Anaerobic digestion process, which involves four major stages of hydrolysis, acidogenesis, acetogenesis and methanogenesis, occurs in the bioreactor. Throughout this process, organic materials are biodegraded into volatile fatty acids (VFAs) and hydrogen as intermediate products and into methane as a final product [2]. All these products can make the AnMBR an energy neutral or even energy positive technology. In addition, unlike anaerobic digestion that requires mesophilic ranges from 35°C to 37°C, AnMBR can be operated in room temperature or even in cold temperatures by expanding SRT, which is both cost and energy-efficient [3].

However, there are some issues that need further attention in AnMBR technology. One of the most challenging issues is membrane fouling, which deteriorates the performance of membrane and shortens membrane lifespan. Membrane fouling generally occurs when the components of sludge interact with membrane material, causing an initial pore blocking and cake layer formation. It is reported that membrane fouling in AnMBR has more severe impacts compared to the aerobic system in terms of pollutant removal efficiency and sludge characteristics, even using the same membrane material [1]. Meng et al. [4] also mentioned that the cake layer formed with anaerobic sludge might have a comparatively lower removability than that with aerobic sludge.

Many researchers have carried out studies on alleviating membrane fouling, which have involved pretreatment of feed wastewater, optimization of operational conditions, membrane module or surface modification [1, 3, 5]. To control membrane fouling, different pretreatment methods have been introduced, such as alkaline pretreatment, acid pretreatment, ozone pretreatment and settling organic contaminants [6-9]. Some studies have shown that feed characteristics have significant impacts on the formation and compactness of the cake layer. Enhancers, such as adsorbent agents, carriers and other chemical agents, can be effective in modifying the properties of feed water in AnMBRs. Adsorbents, such as activated carbon, zeolite and bentonite, promote adsorption and ion exchange phenomena. Coagulants including polyaluminum chloride and ferric chloride, and suspended carriers can promote coagulation and flocculation, respectively. Consequently, enlarged floc size and decreased soluble organics in the supernatant can mitigate membrane fouling [1, 10].

To date, there are many investigations of fouling control in aerobic MBRs by adding different additives as adsorbents [11, 12], coagulants/flocculants [13-16] and suspended carriers [17]. However, only a limited amount of research is available to investigate the effects of enhancers on fouling control in anaerobic MBRs. Additionally, most review papers of fouling control in AnMBRs have only focused on controlling operating conditions and overall mitigation strategies [1, 5, 18, 19]. This is the first review article that focuses on the different kinds of fouling reduction enhancers in AnMBR and their influences on membrane fouling reduction. Effects of adding activated carbon including powdered activated carbon (PAC) and granular activated carbon (GAC) on AnMBR performance will firstly be discussed. Then the following sub-sections will concentrate on the effects of adding other enhancers.

2. Enhancers to AnMBR

2.1 Activated carbon

Activated carbon (AC) has been widely applied in membrane bioreactors (MBRs) due to its high adsorption capability, enhancement of biodegradation, and subsequent removal of recalcitrant pollutants. The addition of AC can also efficiently mitigate membrane fouling, as it has high potential to enhance membrane flux as well as removal performance of chemical oxygen demand (COD) and recalcitrant pollutants [20, 21]. Moreover, the use of AC in the anaerobic digestion process has been gaining more attention because it facilitated the alleviation of organic shock loading, the enrichment of essential anaerobic microorganisms and the improvement of anaerobic digestion stability [21].

There are two types of AC, namely PAC and GAC. PAC has high porosity and large surface area, which can lead to high adsorption capacity, while removing odour, colour and taste [21]. Compared to PAC, GAC has larger size than PAC, which is more easily retained in the reactor and more economical when it is used continuously, because GAC can be regenerated by thermal process. Due to the larger size, GAC also has stronger physical interactions with the membrane surface [22].

2.1.1 Powdered activated carbon

PAC can reduce membrane fouling through three mechanisms. Firstly, at the initial stage of PAC addition, adsorption of organic matters occurs, which greatly removes the dissolved organic matters (DOM) from wastewater. Secondly, after initially adsorbing organic matters and becoming saturated, microorganisms aggregate on the porous surfaces of PAC particles as supporting medium for attached bacterial growth [23]. Finally, the formation of biologically activated carbon (BAC) promotes the degradation of pollutants and modifies the sludge properties, which is the most important mechanism. After the colonization of microorganisms, the planktonic microorganisms transform into biofilm. Once attached bacteria produce extracellular polymeric substance (EPS), it helps not only the attachment of microorganisms to biofilm, but also the stabilisation of the biofilm structure. The active biofilm continues to biodegrade organic compounds as well as to reduce the attachment of microorganisms on the membrane surface so that it can relieve the membrane biofouling [12, 24]. In addition, the scouring effect of PAC also alleviates membrane fouling, as it can remove the deposited cake layer on membrane surface while limiting the accumulation of foulants [25]. Figure 1 illustrates the mechanism of fouling reduction when activated carbon is added.

More recent studies have confirmed that PAC has positive effects on sludge morphology, aggregation ability of sludge flocs and microbial properties, and thus the pollutants removal mechanism can be enhanced. The study by Zhang et al. [26] reported that PAC addition in a submerged AnMBR was able to form larger floc size of the sludge compared to AnMBR without PAC. However, in case of long-term operation over 140 days, the sludge diameter decreased from 20.66 μm to 17.00 μm, which was contrary to the result from previous research about PAC in AnMBR [27]. This may have resulted from the fact that PAC enabled the generation of free living filamentous microbes after the long term operation and prevented the large floc size formation in mixed liquor. In addition, the sludge aggregation ability was able to be assessed using the total interaction energy which is a function of separation distance between the sludge surfaces. It could be calculated by summing Lifshitz-van der Waals energy, Lewis acid-base energy and repulsive or attractive electrostatic double layer energy [28, 29]. PAC addition showed highly negative value of total interaction energy per unit area of sludge, which indicated that the characteristics of sludge surface has transformed into hydrophobicity, and thus sludge cells adherence could be strengthened by stronger attractive interaction. This improvement of aggregation ability led to more stable sludge flocs and potentially reduced the EPS release, which mitigated the pore blocking and irremovable membrane fouling [11]. The increased bacterial diversity and evolution of the bacterial community were also attributed to PAC addition creating additional microbial environment in the form of BAC, which promoted the enrichment and growth of some special functional bacteria. The enrichment of *Acinetobacter, Comamonas, Flavobacterium* and *Pseudomonas*, which can contribute to formation of sludge flocs and degradation of organics, were highly promoted [28]. When PAC was applied in anaerobic batch biofilm reactors as a biofilm carrier for the enhancement of refractory compounds degradation, it increased the abundance of *Methanothrix,* *Methanomassiliicoccus,* and *Methanobacterium* which were favourable for the methane production [30].

In a study of Hu and Stukey [31], 1.7 g/L of PAC addition to submerged AnMBR showed significant benefits for the removal of fine colloidal particles as well as membrane flux improvement and transmembrane pressure (TMP) reduction. It was also found that PAC addition showed 22.4% of increase rate in COD removal, while GAC showed no significant increase. The reason why PAC was more efficient than GAC in terms of COD removal, might be due to the greater surface area per mass than GAC. Another study, which applied 1.7 g/L of PAC as well, showed 30% increased dissolved organic carbon (DOC) removal and decrease in SMP concentration [32]. On the other hand, when the more PAC concentration of 4 g/L was added, the SMP was rather accumulated while having excellent COD and colour removal efficiency [33, 34]. Several studies have revealed that the PAC addition not only decreased turbidity and colour, but also removed potential foulants such as fine colloids and soluble microbial products (SMP), which could lead to the reduction of fouling layer thickness [25, 26, 35]. Table 1 summarizes the effects of PAC addition on membrane fouling control in submerged AnMBRs.

PAC can also be beneficial to AnMBRs in terms of the removal of antibiotics, which have negative impact on membrane fouling. In fact, the existence of antibiotics in AnMBRs has worsened performance and issues associated with membrane fouling, since it could lead to a decrease of the floc size and pH, and an increase in the secretion of EPS and SMP. Moreover, it could facilitate the development of microbial communities which had the most contribution to membrane fouling, such as *Firmicutes*, *Proteobacteria* and *Chloroflexi* [36, 37]. According to a review by Cheng et al. [38], the addition of antibiotics and combined antibiotics to anaerobic reactors, such as sulfamethoxazole (SMX), tetracycline (TC), erythromycin-tetracycline-sulfamethoxazole (ETS), and erythromycin-tetracycline (ST), could cause negative effects on pH, COD removal efficiency, and biogas production. Both the pH value and COD removal efficiency in anaerobic sequencing batch reactors significantly decreased when the high concentration of antibiotics, such as 45 mg/L of SMX, 8.5 mg/L of TC, and 46 mg/L of ETS, were added [39-41]. Biogas generation, which is inherently related to COD removals under anaerobic conditions, was inhibited as well, and the reason for this might be the methanogenesis process was sensitive to the presence of antibiotics in anaerobic processes. Likewise, 100 μg/L of SMX and TC in anaerobic/aerobic-MBR accelerated the rate of TMP rise and decreased the membrane fouling cycle from 25 days to 8 days. In addition, the fouling layer became denser and thicker with 20 μm of thickness. Furthermore, higher concentration of two antibiotics, 1000 μg/L of SMX and TC, resulted in further decrease in membrane fouling cycle to 4 days and 40 μm of the fouling layer thickness [42]. However, the addition of activated carbon could remediate these negative results. As a case in point, the addition of PAC into AnMBR increased removal efficiencies of five different pharmaceuticals including SMX, trimethoprim (Tmp), carbamazepine (Cbz), diclofenac (Dcf), and triclosan (Tcs) by approximately 5%-92%, as the adsorption of pharmaceuticals to PAC thermodynamically enhanced their biotransformation [43].

Although the optimal dosage of PAC resulted in significant alleviation in membrane fouling, overdosing might have contrary results due to its potential to become a foulant. Akram and Stuckey [44] proposed that appropriate amount of PAC should be added for the best improvement of performance of AnMBR and membrane fouling amelioration. In their research, PAC concentration of 1.67 g/L highly improved the flux by more than four times, while 3.4 g/L of PAC caused a decrease of flux and adsorption incapacity of PAC to higher concentration of biomass. The excessive dosage of PAC could result in poor membrane filtration due to the increased sludge viscosity caused by the presence of more extracellular polymers. Moreover, small PAC particles (8-35 µm) at high concentration in suspension increased turbidity of mixed liquor and caused more membrane pore blockage and abrasion [20]. Therefore, the optimal PAC dosage is effective for flux improvement and adsorption of fine solutes, and regular replacement of aged PAC with fresh PAC is necessary [4].

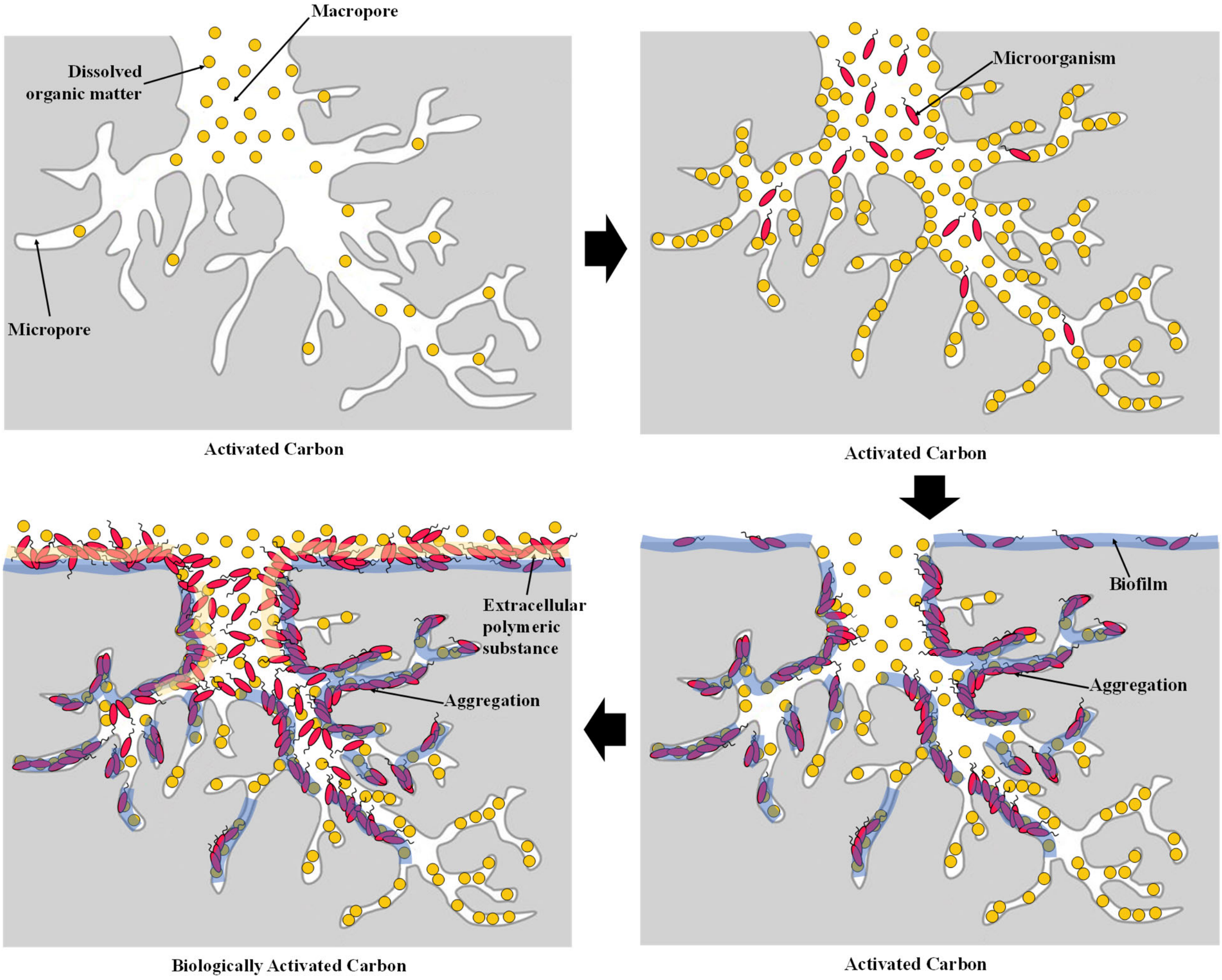

Figure 1. Mechanism of fouling reduction performance of activated carbon

Table 1. Effects of PAC addition on performance of submerged AnMBRs

AC supplier	Dose of AC	Feed water	Operating conditions	Effects on performance[a]	References
• Norits-Super • Total surface area of 1300 m^2/g	1.7 g/L	Saline sewage	• SRT : 250 d • HRT : 8, 20 d • OLR : 2 gCOD/L·d • Flux : 5 - 8 L/m^2·h • Salinity : 0-35 gNaCl/L • Temperature : 35 ± 1°C	• Decrease in TMP by 0.070 bar • Increase in dissolved organic carbon (DOC) removal by 30% in the reactor and 5% in effluent • Reduction of high MW compounds by 70% • Reduction of large flocs attached to the biofilm • Decrease in SMP	[32]
• Norit, Singapore • BET surface area of 925 m^2/g • average particle size of 22 µm	1 g/L	Synthetic sewage	• SRT : 213 d • HRT : 6 h • Flux : 5 L/m^2·h • Feed COD : ~500 mg/L • Total nitrogen : ~100 mg/L • Temperature : 35°C	• Enhanced removal of five selected pharmaceuticals (Tmp, Smx, Cbz, Dcf, Tcs) by about 5-92% • Increased biotransformation of Tmp by 4.5%, Smx by 18.8% and Tcs by 34.8%	[43]
• Norit, UK • Average particle size : 15-25 µm	400 mg/L	Synthetic sewage	• Flux : 15 L/m^2·h • Feed COD : 500 mg/L • VSS : 5.0-5.5 g/L • Temperature : 35°C • pH : 6 - 7	• Reduction in supernatant supra-colloidal particles, colloids and SMPs • Reduced thickness of fouling layer reduced • Declined levels of COD by 13% and proteins	[26]
• Norit, UK	1.67, 3.4 g/L	Synthetic wastewater	• SRT : 250 d • HRT : 6 h • OLR : 16 gCOD/L·d • Feed COD : 4 g/L • Temperature : 35 ± 1°C	• Enhanced performance during start-up period (i.e. shortened start-up duration, increased COD removal, declined SMP level,	[44]

			- Neutral pH	increased concentration of biomass and enrichment of microorganisms) - Adsorption of biodegradable low and high MW residual COD by PAC - Adsorption of fine colloids and dissolved organics by PAC - Improvement in flux (i.e, increase in flux from 2 to 9 $L/m^2 \cdot h$ with 1.67 g/L PAC)	
- Synth®	4 g/L	Textile wastewater	- HRT : 24 h - Temperature : 35°C - pH : 6.8 – 7.2 - Feed COD : 670 mg/L - COD:N:P : 350:5:1	- Enhanced removals of COD, VFA, turbidity and colour by about 11%, 8%, 43% and 69%, respectively - Increased reactor stability - Enhanced membrane permeability (higher critical flux) - Adsorption of toxic compounds, aromatic amines and VFA by PAC - Increased accumulation of SMP	[34, 35]
- Synth®	4 g/L	Domestic sewage	- HRT : 24 d - COD:N:P : 350:5:1 - OLR : 0.53 $kg/m^3 \cdot d$ - Temperature : 35°C - pH : 6.5 – 7.5	- Enhanced COD and colour removal by about 9% and 6.7-12.7%, respectively - Increased reactor stability - Less accumulation of VFA - Adsorption of aromatic amines by PAC	[33]
- Extra pure charcoal powdered activated carbon	1, 3, 5 g/L	Palm Oil Mill Effluent (POME)	- SRT : 30 d - HRT : 6 d - Feed COD: 4.74 ± 1 g/L - Temperature : 35°C - pH : 7-8	- Increased COD removal efficiency at higher PAC dosage - Increased floc size at higher PAC dosage	[25]

		• At mesophilic condition	• More reduction of EPS concentration and membrane fouling at higher PAC dosage	

[a] Cbz, carbamazepine; Dcf, diclofenac; Smx, sulfamethoxazole; TCS, triclosans; Tmp, Trimethoprim

2.1.2 Granular activated carbon

In recent years, the addition of GAC has been extensively applied in anaerobic digestion process to enhance both reactor efficiency and abundance of special functional microorganisms. Anaerobic digestion process contains electron exchange between fermentative bacteria and methanogens in the form of metabolites, such as acetate, H_2 and methanol. Previous studies have demonstrated that conductive additives like GAC enabled direct electron exchange instead of metabolites, which could eventually enhance methanogenic conversion of short-chain fatty acids, such as acetate, butyrate, and propionate, and subsequent improvement of methane production [45-47]. Zhang et al. [48] showed that GAC remarkably promoted methanogenesis by enhancing direct interspecies electron transfer between fermentative bacteria, *Geobacteraceae*, and methanogens, *Methanosaetaceae.* Another research has also concluded that surface modified GAC with magnetite stimulated enrichment of electroactive bacteria, such as *Shewanella, Pseudomonas, Geobacter and Desulfuromonas*, enhancing the methane production by a degradation of propionate to acetates and electrons that can be utilized by methanogens [45].

As a membrane fouling mitigation strategy, methods of inducing unsteady-state shear on the membrane surface, such as bubbling and vibration, have been applied to MBRs. Particle fluidization recently has been presented as an alternative to bubbling, as it could have the same effect on membrane fouling reduction with at least ten times lower energy requirements than bubbling. Particularly, the fluidization of GAC has gained significant attention, because larger GAC was more effective during long-term operation [49, 50]. Therefore, previous studies have reported that GAC fluidization resulted in significant membrane fouling alleviation in anaerobic fluidized membrane bioreactor [51-56]. In the studies of integrated anaerobic fluidized-bed membrane bioreactor, high amount of protein was adsorbed resulting in remarkable improvement of membrane filtration [52, 53]. The effect of GAC fluidization has been demonstrated with its energy efficient and effective advantages, unlike the popular air-sparging method which required comparatively high energy costs [57]. As GAC fluidization is one way to induce unsteady-state shear on membrane, which has been identified as a cost-effective method, it could reduce the energy requirement in the process [58]. As a case in point, the electrical energy requirement for anaerobic fluidized-bed ceramic membrane bioreactor operation was estimated to be 0.039 kWh/m^3, which was only 17% of electrical energy that can be generated from produced methane [59]. The average energy consumption of GAC fluidization is generally reported as 0.15 kWh/m^3, whereas that of gas sparging is twice higher, which is 0.31 kWh/m^3 including pumping and mixing [60]. The table 2 summarizes the effects of GAC in anaerobic fluidized membrane bioreactor.

In the research of Ding et al. [61], they added 50 g/L of GAC to an expanded membrane-coupled granular sludge bed (EGSB) and showed a remarkable enhancement of COD removals (80% vs 62% without PAC) and decrease in SMP concentration. The cake layer resistance, which was the main fouling mechanism in membrane-coupled EGSB process, was also decreased by 53.5%. Another research by Wang et al. [62] treating wastewater containing phenol and quinolone reported that a 2 g/L of GAC could not only remove COD and SMP by adsorption, but also enhance the degradation of phenol and quinolone. The high adsorption capacity of GAC could capture some fouling-causing compounds like SMP prior to attachment on membrane surface. Meanwhile, GAC could scour the foulants from the membrane surface and prevent the accumulation of foulants. Hence, the use of GAC as suspended medium effectively mitigated irreversible fouling [63]. As a case in point, the scouring effect of GAC with flux of 16 L/m^2·h in a two-stage anaerobic fluidized membrane bioreactor was able to mitigate membrane fouling, along with effective removal of 20 commonly found pharmaceuticals (i.e. ibuprofen, caffeine, and SMX, etc.) through adsorption and biodegradation [64, 65].

Although having positive effects on membrane fouling control, Wu et al. [55] suggested that the behaviour and characteristics of GAC might have a harmful influence on membrane performance. Due to the fine carbon particles that are released from GAC itself during fluidization, the fouling could be aggravated by blocking the pores and forming a thin cake layer. GAC abrasion also led to a partial loss from the initial membrane quality. The reduction in adsorption capacity of GAC over time was also a major limitation. Thus, the exhausted GAC needs to be replaced or regenerated by thermal process to recover the adsorption capability [24].

Furthermore, the energy requirements for fluidization and membrane fouling mitigation were significantly different depending on the particle size of GAC as well as adsorption capacity. When the adsorption of fresh GAC predominantly took place, comparatively small GAC particles had greater effect on fouling reduction due to large surface area, along with less energy consumption. However, after the adsorption capacity was exhausted, dominant process for fouling reduction became the scouring effect. Then, the relatively large GAC particles were more effective in fouling reduction, but more energy is required for fluidization [66]. Charfi et al. [67] showed that 2-3 mm of GAC particles acted as a better method of fouling reduction by removing cake layer on membrane surface, whereas small particles from 0.18 mm to 0.5 mm rather intended to accumulate on membrane surface. It was also found that, although large particles were more effective in scouring due to inertial forces, the energy requirement on fluidizing the particle was also higher [49]. Thus, further studies are necessary for a better understanding of production of fine carbon particles, GAC abrasion and choosing suitable particle sizes for mitigating membrane fouling.

rmance of fluidized AnMBRs

	Feed water	Operating condition	Effects of GAC on performance	References
	Primary-settled domestic wastewater	• SRT : 485 d • HRT : 4.5 – 6.8 h • Temperature : 8 - 30 ℃ • Average effluent COD : ~23 mg/L	• No chemical membrane cleaning	[54]
	Synthetic wastewater	• HRT : 8.7 h • Feed COD: 150 mg/L • Flux : 5 $L/m^2 \cdot h$ • Temperature : 25 ℃	• Complete removals of diclofenac, ibuprofen and sulfamethoxazole	[64]
n	Municipal wastewater primary-clarifier effluent	• HRT : 2.3 h • Flux : 6 - 11 $L/m^2 \cdot h$ • OLR : 1.0 – 3.5 $kgCOD/m^3 \cdot d$ • Temperature : 25 ℃	• No requirement of other fouling control process • Lower electrical energy requirements	[56]
n	Domestic wastewater	• HRT : 1.3 – 2.1 h • Feed COD: 250 mg/L • Flux : 22 $L/m^2 \cdot h$ • Temperature : 25 ℃ • pH : 7.3 -7.6	• Lower biosolids production • Low energy requirement (10% of energy converted form methane)	[51]
	Dilute wastewater	• HRT : 1.3 – 2.1 h • Feed COD: 300mg/L • Flux : 22 $L/m^2 \cdot h$	• Lower energy requirement • No adverse effect of	[59]

Two-stage AnFMBR	• 10 × 30 mesh • Bulk density: 500–1000 m^2/g • specific gravity: 0.85 and 2 g/cm^3 • 25% in AFBR, 50% in AFMBR	Municipal wastewater	• HRT : 1.28 h • OLR : 5.65 $kgCOD/m^3 \cdot d$ • Feed COD: 250mg/L	• Effective removals of pharmaceuticals • No requirement of other fouling control process	[65]
AnFMBR	• >2mm, • 0.85-2mm • 0.5-0.85mm • 0.18-0.5mm • <0.18mm • 10%, 30%, 50%	Synthetic wastewater	• Flux : 50 $L/m^2 \cdot h$ • Feed COD: 250 mg/L	• Energy requirement increased with particle size • The higher the packing ratio, the greater the fouling reduction	[66]
AnFMBR	• N/A	Screened domestic wastewater	• SRT : 12 d • HRT : 4 h • Flux : 7.6- 7.9 $L/m^2 \cdot h$ • OLR : 1.3 -1.4 $kgCOD/m^3 \cdot d$ • Temperature : 13 - 32°C	• Similar BOD_5 and COD removal efficiencies (around 85%) achieved when operating at a 65% shorter HRT than gas-sparing system (removals of around 90%)	[60]
AnFMBR	• Lignite coal (0.42-0.85mm, 650 m^2/g) • Peat bog (0.85-2.4mm, 600-800 m^2/g)	Synthetic wastewater	• Flux : 14 $L/m^2 \cdot h$	• Fouling reduction and detrimental effects on membrane	[55]

	• Peat bog (2.4-4.6mm, 600-800 m^2/g)				
IAFMBR	• 40g • 10 × 30 mesh	Domestic wastewater	• HRT : 6 h • Flux : 7.1 $L/m^2 \cdot h$ • Feed COD: 247 - 449 mg/L • Permeation : 23.2 L/d • Temperature : 35, 25, 15 ℃ • pH : 7.18 – 7.99	• High protein adsorption by GAC	[53]
IAFMBR	• 200 -300g • 10 × 30 mesh	Domestic wastewater	• HRT : 4, 6, 8 h • Flux : 0.27 $m^3/m^2 \cdot d$ • Feed COD: 300 mg/L • Temperature : 35 ± 2℃ • pH : 7.5 ± 0.21	• Adsorption of protein in cake layer by GAC • Improved membrane filtration	[52]

[a] AFCMBR, anaerobic fluidized bed ceramic membrane bioreactor; AnFMBR, anaerobic fluidized membrane bioreactor; SAF-MBR, staged anaerobic fluidized membrane bioreactor; IAFMBR, integrated anaerobic fluidized-bed membrane bioreactor; SAF-CMBR, staged anaerobic fluidized bed ceramic membrane bioreactor.

2.2 Other enhancers

2.2.1 Biochar

Biochar is a porous and carbonaceous residue obtained from thermal decomposition of biomass in an oxygen deleted environment, or from other processes such as pyrolysis, hydrothermal carbonisation, gasification and torrefaction. It is usually produced at a lower temperature than 700 °C, because reaction above 900 °C causes the destruction of walls between pores, which results in widening of pores of biochar [68, 69]. Unlike AC, it is produced without any activation, and this non-activation makes the specific surface area of biochar less efficient compared to AC. However, the production cost of biochar is one tenth cheaper than that of AC [68, 69, 71]. After a series of reactions of biomass such as dehydration, depolymerisation and carbonisation during thermal decomposition, three products, namely condensable liquid (bio-oil), non-condensable gases (syngas) and biochar are produced, which depends on the type of biomass used and process conditions (i.e. temperature and residence time). Biochar usually consists of fixed carbon, labile carbon and other volatile compounds, as well as moisture and ash. Fast pyrolysis aims at liquid oil production, whereas the goal of slow pyrolysis is biochar production, as the slow evaporation of water and release of volatile components can result in an increase in relatively fixed carbon content of the solid [70, 72]. The heterogeneous surface of biochar, which has both carbonised and non-carbonised fractions, accommodates several adsorption mechanisms. Physical adsorption, surface precipitation and the pore-filling are the major routes of adsorption. Moreover, for positively charged organic compounds, hydrophobic effect and hydrogen bonding of biochar surface are the important adsorption routes. On the other hand, the removal of inorganic compounds largely depends on electrostatic attraction, precipitation and ion exchange [70, 73].

The addition of biochar on anaerobic digestion process has shown to be effective in terms of biogas production and selectively enriched microbial groups. The biochar addition was able to enhance VFA production and degradation, and improve both hydrogen and methane production [68, 74]. Some studies demonstrated that better biogas production could be obtained from enhanced direct interspecies electron transfer (DIET) process by enrichment of electrogenic *Geobacter* and *Bacteroidetes*, which are potential direct interspecies electron transfer partners during anaerobic digestion [75, 76]. In addition, since biochar contains redox active moieties such as quinines, phenolics and phenazines, they can catalyse the electron transfer between biochar and outer membrane cytochromes during redox reactions [77, 78]. Moreover, biochar addition significantly enhanced methanogenesis by facilitating the enrichment of *Methanosarcina* even in high ammonium stress, and also favoured anaerobic sludge granulation, due to the ability of promoting biofilm formation and reducing the inhibition behaviour of ammonia [79, 80]. Similar to the biogas yield enhancement, biochar could facilitate hydrogen production via enrichment of hydrogen-producing bacteria [81]. The alleviation of sulphide toxicity during anaerobic treatment of sulphate-rich wastewater using biochar was investigated and biochar promoted reactor stability by adsorption of H_2S from biogas [82]. Due to the adsorption capability and functional groups on the surface, biochar was also able to adsorb EPS and enhance sludge granulation, which could lead to significant mitigation of membrane fouling in aerobic MBRs [83-85].

Some previous studies have demonstrated that biochar could be beneficial to anaerobic membrane bioreactor. Bamboo charcoal, one kind of biochar, was able to enhance the removal performance of AnMBR as well as mitigate membrane fouling. In this study, two AnMBRs treating bamboo industry wastewater were analysed with and without bamboo charcoal addition. The result showed that COD removal efficiency increased about 5% after the addition of bamboo charcoal, as well as reduced membrane fouling owing to the decrease of both SMP concentration and resistance of the fouling layer. Meanwhile, the methane yield became higher as a result of greater microbial activity of dominant microorganisms in methane production, such as *Methanosaeta, Methanospirillum* and *Methanobacterium*, occurring additionally inside the pores of the bamboo charcoal [86]. More recent study showed that membrane fouling was effectively reduced in a biochar-amended AnMBR along with 56% of decreased TMP rising rate and decreased proteins of EPS. In addition, *Arcobacter*, one of the bio-foulants that is involved in membrane biofouling, was hardly accumulated due to the presence of biochar [87].

2.2.2 Waste yeast

Waste yeast is traditionally used as a protein supplement in animal feed or alimentary substrate for the food processing industry. The brewing industry is the major source of spent yeast, which also produces other residues in addition to brewery wastewater, such as methanogens and small cellulosic particles [88]. Due to the high degradation capacity of yeast, it was favourable in treatment of landfill leachate, which contained high amount of recalcitrant compounds like phenolic compounds as well as toxic substances such as halogenated and heavy metals [89]. Yeast had lower tendency to adhere on membrane surfaces than other microorganisms, so that its application in MBR can be beneficial in membrane fouling control and system operation [89]. The presence of yeast in aerobic MBR could significantly remove not only COD, colour and EPS, but also refractory substances including polyacrylamide [90-93].

Anaerobic co-digestion, which balances the nutrient component of different residues, is widely applied as a way to dilute potential toxic compounds and enhance biogas production [94]. Some previous studies have demonstrated that additional co-substrates in anaerobic wastewater treatment increased methane production by maintaining a pH level within the methanogenesis range between 7.0 and 7.5, while improving the degradation of low biodegradable substrates [88, 95-97]. The research on the supplementation of yeast in brewery wastewater treatment as a co-substrate in a upflow anaerobic sludge blanket (UASB) reactor showed enhanced biogas production by 50%, while no significant changes in COD removal efficiency and accumulation of VFAs were observed up to 1.1 (v/v)% of brewery yeast concentration [88, 94]. These results indicated that the additional waste yeast could be a feasible substrate in anaerobic digestion in terms of high biodegradability and biogas yield.

The supplementation of yeast wastes as co-substrate in an AnMBR can have positive effects on membrane fouling control as well. A research by Yun et al. [98] investigated the effects of yeast on AnMBR performance treating low-grade coal wastewater (LCWW). Compared to no methane production in the absence of yeast wastes, AnMBR with yeast wastes gradually increased COD removal efficiency as well as methane production. In addition, the presence of yeast wastes showed the significant growth of some microorganisms such as *Methanococcus* and *Methanosarcina* which were responsible for the degradation of LCWW and biogas production. However, due to the metabolism of these bacteria, the fraction of SMPs and aromatic group with high molecular weight (> 1 kDa) also increased. Thus, the addition of yeast wastes could be a potential alternative as an additive to AnMBR due to their positive effects on biodegradation of LCWW and growth of microorganisms, but further research should be carried out to find out the effect on fouling control.

2.2.3 Iron

Iron, which is the most abundant transition metal on the Earth, is also an essential component for the growth of most living organisms. Although iron itself is a non-toxic and electron donor in redox reactions, the presence of iron in anaerobic environment plays an important role in the electron cycling and metabolic activity of microorganisms [99, 100]. The Fe(II) and Fe(III), which are generated from several iron compounds, can be provided as nutrients for microbial activity or as redox mediators to facilitate the conversion of organic matters to methane [100]. One of the strong reductants, zero-valent iron (ZVI), is an active anode material in electrocoagulation, and electrically produces Fe(II) ions which promote coagulation and effectively decrease the soluble and colloidal particular matters. Although the generation of Fe(II) ions from ZVI in aerobic process needs to be triggered by the electric field, it can occur spontaneously in the anaerobic digestion process. The protons, which are released by acidogens during the acidification, can help spontaneous generation of Fe(II) ions without any drive of electric field [101].

Many previous studies have investigated the addition of iron or ZVI into anaerobic digestion which could significantly increase methane production and improve COD removal [102-105]. When iron was added to anaerobic aquatic environment, hydrogen was produced by iron corrosion. This hydrogen evolution can benefit methane production by enhancing both hydrogenotrophic methanogenesis and homoacetogenesis. In addition, iron was able to serve as an electron donor to reduce oxidation-reduction potential (ORP), and led to the decrease in propionic-type fermentation and subsequent enhancement of methanogenesis. As the accumulation of propionate destroyed the pH balance between acidogenesis and methanogenesis as well as hindered the methanogenesis of acetate, it should be reduced during the anaerobic process [106, 107]. Although anaerobic digestion can be limited by low efficiency of hydrolysis and acidification, ZVI could intensify the activities of enzymes related to hydrolysis and acidification, such as protease which is responsible for catalysing hydrolysis of polysaccaharide to monoses [107]. Moreover, the presence of ZVI stimulated the growth of hydrogen-consuming microorganisms such as homoacetogens and hydrogenotrophic methanogens, thereby enhancing acetate or methane production [107, 108]. In addition, iron could also effectively eliminate odorous H_2S gas by precipitation of FeS. Likewise, the iron might be used for phosphate recovery in the form of compounds of iron and phosphate such as vivianite [99]. It was also found that the supplementation of iron salts to anaerobic digestion could be potentially advantageous to membrane fouling by supporting granulation and stabilisation. When ferrous iron was supplied to UASB reactor, anaerobic bacteria and EPS tended to adhere to iron in order to form a more stable structure with 56% of enlarged granule diameter. Moreover, inorganic precipitates such as ferrous sulphide could contribute to the stability of granules [109, 110].

The iron addition to AnMBR provided remarkable benefits on membrane fouling mitigation in several previous studies. Dong et al. [111] showed the influence of $FeCl_3$ as an additive in long-term operation of an AnMBR treating municipal sewage. The performance of the AnMBR, including removal efficiencies of COD and BOD_5, was enhanced by adding 26 mg/L of $FeCl_3$. Furthermore, even though the addition of $FeCl_3$ caused the increase of mixed liquor suspended solids (MLSS) concentration and formation of a more thickened cake layer, membrane fouling has been mitigated due to the more porous cake layer formation and increased filterability of mixed liquor. Zhang et al. [26] also investigated the addition of $FeCl_3$ to AnMBR, which effectively reduced membrane fouling by increasing both sludge floc size and the colloids as well as decreasing SMPs. Since iron remained in the reactor as a precipitate, resulting in minimal concentration of iron in the effluent or supernatant, it was expected to be advantageous for a long term operation. In a recent study, ZVI has been applied into AnMBR with and without electric field. Although ZVI with electric field facilitated the increase of iron releasing rate of ZVI by 12 times and enhanced removal performances of COD and total phosphorous (TP) by about 3% and 50%, respectively, it resulted in more severe fouling due to the high density of Fe-rich fouling layer. However, ZVI without electric field significantly mitigated membrane fouling rate by 20% through the enhancement of mixed liquor filterability [101].

2.2.4 Calcium

Calcium can be another special additive to alleviate fouling and enhance characteristics of granular sludge by enhancing bioflocculation. EPS, which is known to be the main substance affecting membrane fouling, typically contains negatively charged functional groups such as hydroxyl and carboxyl. Due to negatively charged EPS, cations play an important role in sludge flocculation. Divalent cations including calcium ions tend to combine preferentially with carboxylic functional groups of EPS and form bridges between the EPS molecules. This bridge formation promotes the improvement of bioflocculation, enlarges flocs and mitigates fouling [112-115]. Since the cost of calcium salts is relatively low, they have been widely used in aerobic process as an additive, which improve the properties of mixed liquor [114, 116]. However, the decline in permeability and subsequent inorganic fouling occurred with high concentration of calcium of 830 mg/L, due to the precipitation of calcium carbonate. Therefore, more research is necessary to have a better understanding of effects of calcium addition and find out the optimal calcium concentration [117].

The addition of calcium can also positively affect anaerobic processes. Some previous studies have been conducted to evaluate the influence of calcium addition and the most effective dosage for anaerobic digestion. When five different concentrations of calcium chloride ($CaCl_2$), which are 0, 1, 3, 5, and 7 g/L, were added to anaerobic digestion process, 3 g/L of calcium concentration was optimal for the best performance of anaerobic digestion and biogas production [118]. Similarly, according to a study of Ahmad et al. [119], calcium oxide (CaO) in the UASB reactor enhanced granulation and the accumulation of biomass as well as the degradation of butyrate and acetate acid. Since the addition of calcium on anaerobic digestion process significantly increased the abundance of *Methanosaeta* as the dominant methanogen, the methane production could be improved [120]. However, an overdose of calcium from 5 to 7 g/L of concentration, which may lead to precipitation and limit mass transfer between microbes and organic compounds, further inhibit anaerobic process. When the precipitates such as calcium carbonate were formed on the surface or within the granules, they can cause sludge washout, as well as the declined methanogenic activity and diffusion limitation [118, 119].

Due to the positive effects of calcium addition on anaerobic process, the use of calcium as an additive in membrane bioreactors can also be a promising way to reduce membrane fouling [121]. An investigation on the effects of calcium addition (0, 50 and 100 mg/L of calcium) was conducted in three sequencing batch reactors with external dead-end microfiltration. The result showed that the highest dosage of calcium was able to enhance the reduction of fine particles, EPS and colloids in supernatant, leading to the mitigation of membrane fouling [122]. This significant reduction of membrane fouling was mainly due to calcium promoted bioflocculation, which achieved high volumetric organic removal and increased methane production rate. Furthermore, the enlarged size of anaerobic sludge granules by calcium addition was also reported in some studies using membrane-coupled expanded granular sludge bed (EGSB) reactor or UASB reactors [123-126]. When calcium chloride was added to the EGSB reactor, the membrane fouling was alleviated effectively and the concentration of SMP decreased [121].

2.2.5 Polyaluminum chloride

One of the aluminum salts, polyaluminum chloride, generally consists of various polynuclear aluminum hydrolysis products, including Al monomers such as $Al(OH)^{2+}$, dimer ($Al_2(OH)_2^{4+}$), trimer ($Al_3(OH)_4^{5+}$), Al_{13} ($AlO_4Al_{12}(OH)_{24}(H_2O)_{12}^{7+}$) and aluminum hydroxide ($Al(OH)_3$). Due to the presence of these products, polyaluminum chloride is superior to the traditional aluminum coagulants, such as $AlCl_3$ and $Al_2(SO_4)_3$, for removing organic matters [127]. The behaviour of aluminum coagulants can be greatly affected by basicity values (B), which is the molar ratio of OH/Al^{3+}, because the dominant hydrolysis products are different under different basicity conditions. Polyaluminum chloride with high basicity value (B = 2.4) resulted in increased membrane fouling propensity, as well as higher DOC removal efficiency and zeta potential of flocs, compared to polyaluminum chloride with lower basicity (B = 2.0 and B = 1.6). This phenomenon might be related to the different dominant mechanisms of coagulation according to the content of Al species. As the percentage of Al_{13} increased along with the basicity value increase, it could provide a larger amount of positive charges for charge neutralization rather than adsorption bridge effect. As the flocs produced from charge neutralization are smaller than those from adsorption bridge effect, it could result in more severe membrane fouling [128].

Many previous studies have reported high charge neutralization capacity of polyaluminum chloride, which can lead to enlarged floc size and better filtration performance. The dose of polyaluminum chloride and subsequent hydrolysis can provide positive charge, which can neutralize the negatively charged sludge flocs and colloids. This neutralization results in weaker repulsion among flocs and colloids, and easier formation of large particles. In addition, SMP and EPS in mixed liquor can be compressed and removed from membrane surface by the charge neutralization and adsorption of polyaluminum chloride [129]. When polyaluminum chloride was added in anaerobic digestion process, it facilitated the reduction of SMP and improved sludge filterability. However, a high concentration of polyaluminum chloride over 500 mg/L in anaerobic digestion could inhibit short chain fatty acids (SCFA) production as well as anaerobic process such as hydrolysis, acidogenesis and methanogenesis by decreasing the ratio of bioavailable nutrient, especially phosphorous [130, 131].

The positive effects of polyaluminum chloride dosing into AnMBR as an inorganic coagulant on membrane fouling control have been studied in some previous research. The addition of polyaluminum chloride could influence microbial characteristics as well as cake layer structure in the anaerobic digestion process. The abundance of anaerobic microorganisms, especially *Cloacimonetes* and *Smithella*, was significantly enriched in AnMBR [132]. Moreover, polyaluminum chloride was able to increase the hydrogen yield by washing out hydrogen consumers, including *Acetoanaerobium* and *Desulfobulbus* [127]. It was also reported that the cake layer on the membrane surface became more porous and looser when polyaluminum chloride was added, which could provide better filterability. A result from a study showed that polyaluminum chloride dosing could lower the composition rate of carbohydrate in SMP and EPS, as well as compress the concentration of EPS. This resulted in reduction of adherence capacity of sludge and more substantially porous cake layer [129, 133].

2.2.6 Zeolite

Zeolite is a porous substance with high crystallinity, which mainly consists of aluminium, oxygen, and metals such as titanium, tin, and zinc. While natural zeolite can be normally found in rocks near volcanoes all over the world, it can also be synthesized or modified in order to improve properties for different applications. Both natural and modified zeolites can be used for adsorption and ion exchange. The presence of cations like Na^+, Ca^{2+}, K^+ and Mg^{2+} on the porous surface of zeolite facilitates ion exchange from a contact solution. Thus, the use of zeolite can be applied in both aerobic and anaerobic biological processes including nitrification and denitrification, activated sludge, and anaerobic digestion. In aerobic processes, zeolite can act as an ion exchanger as well as a biomass carrier, whilst in anaerobic processes, zeolite can also act as an inhibitor of ammonia and heavy metals by ion exchange [134-136].

Zeolite has been reported to improve the performance of anaerobic processes as a porous microbial carrier as well as ion-exchanger. Its high ion-exchange capacity can contribute to enhance NH_4^+ removal which is known as an inhibitor of anaerobic digestion. Indeed, Lin et al. [137] used modified zeolite to reduce NH_4^+ concentration by ion exchange with Na^+ and Ca^{2+} as dominant ions for NH_4^+ adsorption. Additionally, the application of zeolite also showed remarkable improvement in methane production and COD removal [138-140]. These improvements of anaerobic digestion can be attributed to ion exchange of NH_4^+, cations like Ca^{2+} and Mg^{2+} and long chain fatty acids (LCFA) [140, 141]. Another researcher focused on the microbial communities apart from ion exchange, and suggested that zeolite could specifically preserve the growth and immobilisation of microorganisms, especially *Methanosarcina* and *Methanobacteriums* [142].

Zeolite has been widely applied to aerobic and anaerobic membrane bioreactor for membrane fouling reduction. This is because it can improve the settlement of sludge as well as the removal of nitrogen and phosphorous. As zeolite has high porosity and large surface area, it provides a stable environment for bacterial attachment, and substantial microbial aggregation can enhance the settleability of sludge [143]. As a result, membrane fouling can be alleviated by forming rigid sludge flocs and enhancing membrane permeability. Likewise, the application of zeolite as a carrier showed effective removal of COD and suspended solid (SS) which could facilitate better membrane performance and less fouling in anaerobic fluidized membrane bioreactor [144, 145]. When an anaerobic fluidized membrane bioreactor was operated with natural zeolites as carriers, the removal rate of SS significantly improved by 22%. It was also observed that the anaerobic microorganisms were able to attach on the surface of zeolites with remarkable growth. Thus, no membrane fouling was observed due to the low COD and SS concentrations [144].

2.2.7 Beads

The use of granular media fluidizing in AnMBR has been gaining attention along with the development of anaerobic fluidized bed membrane bioreactor. Previous studies showed that fluidization of glass beads and polyethylene terephthalate (PET) beads can act as turbulence promotors and scouring media, respectively, which effectively controlled membrane fouling. Polymer-based gel beads have also been proved to be an ideal microbial carrier, based on their cost effectiveness, high bio-compatibility, strong stability for long-term use, as well as porous structure for microbial attachment and aggregation. Moreover, controlling the size and density of the beads can be achievable by changing the synthesis conditions [146, 147]. Polyvinyl alcohol (PVA), which is water soluble polymer, can form gel beads by cross-linking with other materials like sodium alginate and chitosan [148]. Moreover, when PVA and chitosan form more stable structure through covalent bonds with metal ions, they can be applied in different fields as adsorbent materials, antibacterial agents, or biocarriers [149, 150]. A research by Wang et al. [146] showed the effect of PVA/chitosan gel beads and PVA/chitosan/iron gel beads on anaerobic sludge. Both gel beads favoured the adhesion and aggregation of methanogens, mainly *Mathanospirillum*, *Methanosaeta* and *Methanobacterium*.

According to Düppenbecker et al. [151], the use of fluidized glass beads in AnMBR with external tubular membrane could be a promising option for alleviating membrane fouling in AnMBRs. The optimal diameter of 1.5-mm fluidized glass beads reduced the fouling despite the membrane has been damaged by abrasion. Moreover, the COD removal rate was remained between 77% and 83%, and methane production increased by around 30% as well. The same research group also evaluated the fouling behaviour by three different ceramic membranes including ZrO_2 and Al_2O_3 ultrafiltration membranes and TiO_2 microfiltration membrane. The presence of fluidized glass beads was able to reduce the fouling rate by around 95% for all three membranes. Although all types of membranes were damaged by abrasion of glass beads, Al_2O_3 microfiltration membrane showed the least abrasion in a clean water filtration test [152]. Similarly, the fluidization of PET beads with bigger size and lower density also demonstrated significant fouling reduction by scouring the membrane [153, 154]. These fluidized beads can mitigate membrane fouling by two mechanisms. Firstly, the mixing action of particles can lead to increase in turbulence and thus concentration polarization can be decreased. Secondly, scouring effect on the previously deposited foulants can also alleviate fouling [151].

Table 3 lists the effects of the above-mentioned enhancers on AnMBR performance and Table 4 summarizes the advantages and disadvantages of different enhancers for fouling reduction.

on AnMBR performance

ose	Configuration of reactor[a]	Feed water	Operating conditions	Effects of other enhancers on performance	References
.01 g/L	External AnMBR	Bamboo industry wastewater	• SRT : 150 d • HRT : 3 d • OLR : 6 kgCOD/m^3·d • Temperature : 32 ± 2 ℃	• Increase in COD removal efficiency by 5% • Increase in biogas production and methane yield • Declined SMP concentration and cake layer resistance • Increased microbial diversity and activity of methanogens	[86]
g/L	Biochar-amended external AnMBR	Pharmaceutical wastewater	• SRT : 120 d • HRT : 24 h • VSS : 14.67 g/L • OLR : 7 kgCOD/m^3·d • Temperature : 32 ± 2 ℃	• Effective removal of adsorbable organic halogen (AOX) (average 61.5% vs 56.2% without biochar) • Decrease in TMP rising rate by 56% • Slower TMP jump • Reduced proteins of EPS • Decrease in abundance of biofoulant, mainly *Arcobacter*	[87]
CWW to W of :50	Submerged AnMBR	Low-grade coal wastewater	• HRT : 1 d • COD : 2 g/L • OLR : 1 kgCOD/m^3·d • Temperature :	• High COD removal efficiency (58%) and methane production (182 CH_4 mL/g COD); • Improved degradation of	[98]

Iron	N/A	26 mg/L of $FeCl_3$	Pilot scale AnMBR	Municipal wastewater	• HRT : 8.5 h • SRT : 70 d • Flux : 17 LMH • COD : 383 ± 113 mg/L • pH : 6.7 – 6.8 • Temperature : 23 ± 1°C	• Improved COD and BOD_5 removals by 13.8% and 10.8%, respectively • Increased filterability of mixed liquor with reduced colloidal matter • Increased porosity of fouling layer	[111]
	N/A	150 mg/L of $FeCl_3$	AnMBR	Synthetic sewage	• Flux : 15 LMH • Feed COD : 500 mg/L • VSS : 5.0-5.5 g/L • Temperature : 35°C • pH : 6 - 7	• Increased sludge floc size and colloids particle size • Reduced thickness of fouling layer • Decreased levels of COD and proteins	[26]
	Two pairs of electrodes with flat ZVI anodes (90 cm × 5 cm × 0.3 cm) and titanium cathodes (90 cm × 5 cm)		AnMBR	Municipal wastewater	• HRT : 10 h • Flux : 15 LMH • Temperature : 35 °C • COD : 483 ± 16 mg/L	• Enhanced COD removal by 3%, and TP removal by 50%, and H_2S removal (> 500 ppm with electric field vs < 60 ppm without electric field) • Membrane fouling mitigation by the improvement of mixed liquor filterability	[101]
Calcium	367.5 mg/L of $CaCl_2$	100 mg/L	Dead-end microfiltration	Granular sludge mixed liquors from SBR	• HRT : 4 h • SRT : 96 d • SS : 5000 mg/L • TMP : 5 kPa	• Reduction of fine particles, colloids and SMP • Limited deposition of fine - particles and colloids on the membrane as cake layer served as a prefilter	[122]

	2.5 mM of $CaCl_2$	100 mg/L	Membrane-coupled EGSB	Synthetic wastewater	• HRT : 4 h • COD : 310 – 360 mg/L • pH : 7.0 – 7.5 • TMP : 30 kPa • Flow rate : 0.75 L/h	• Decrease in SMP concentration in the effluent by 47.7 - 60.7% • Decline in cake layer resistance by 42.8% • Delayed transition from pore blocking to cake filtration	[121]
Polyaluminum chloride	N/A	500 mg/L	AnMBR	Anaerobic sludge	• HRT : 3.4 d • SRT : 40 d • TS : 30.16 g/L • VS : 1.2 g/L • Temperature : 35 °C	• Improved filterability of mixed sludge liquor • Decreased concentration of SMP and zeta potential of sludge • Reduced TMP increase rate	[130]
	N/A	200 mg/L	AnCMBR	Phenol- and quinoline-containing wastewater	• HRT : 48 h • Flux : 4.32 LMH • MLSS : 35.77 g/L • MLVSS : 20 g/L • Temperature : 35 ± 1°C	• Changes in the structure of bulk sludge and cake layer • Changes in the component of SMP and EPS • Reduced specific resistance to sludge filtration	[129]
	N/A	500 mg/L	AnMBR	Excess anaerobic sludge	• HRT : 3.4 d • SRT : 40 d • TS : 30.16 g/L • VS : 13.22 g/L • Temperature : 35 °C	• Enrichment of anaerobic microorganisms such as *Cloacimonetes* and *Smithella* • Slightly reduced ratio of VS/TS	[132]
Zeolite	Natural zeolite (0.2 – 1 nm of pore size, 3 of hardness, 2.1 g/cm^3 of density)	350 mL	AnFMBR	Campus domestic wastewater	• HRT : 2.5 h • COD : 130 ± 38 mg/L • pH : 7.2 ± 0.2 • Flux : 10 LMH	• Increase in SS removal rate by 22 % • TMP < 0.2 bar • Significant growth of anaerobic microbes on the surface of zeolite	[144]

					- Temperature : 20 - 35 °C		
Beads	Glass beads (soda-lime glass, 2500 kg/m^3 of density, Worf Glaskugeln, Germany)	4 mm as support layer	AnFMBR	Municipal wastewater	- HRT : 1.7 h - SRT : 46 d - COD : 369 ± 98 mg/L - pH : 7.0 – 7.5 - TMP : 30 kPa - Temperature : 20 °C - Upflow velocity : 24 – 37 m/h	- Increase in methane production by around 30% - Reduction of the fouling rate by around 95% - Least abrasion by Al_2O_3 microfiltration membrane	[151, 152]
	Polyethylene terephthalate beads (3 mm of size and 1.3 of specific gravity)	0.4 v/v of packing ratio	AnFMBR	Synthetic wastewater	- HRT : 3.75 h - SRT : 37.5 d - COD : 250 mg/L - pH : 7.0 – 7.5 - Temperature : 25 °C - Flux : 10 LMH	- Higher effectiveness when applying PET beads with bigger size and lower density - Significant fouling mitigation by souring	[153]

[a] AnCMBR, anaerobic ceramic membrane bioreactor; AnFMBR, anaerobic fluidized membrane bioreactor; AnMBR, anaerobic membrane bioreactor; EGSB, expanded granular sludge bed;

Table 2. Advantages and disadvantages of fouling reduction enhancers

Enhancers	Advantages	Disadvantages
PAC	• High adsorption capacity • Increase removal efficiency of COD, fine colloids, colour, and antibiotics • Improve aggregation ability leading to stable sludge flocs • Large surface area than GAC	• Decrease sludge particl long-term operation • Overdosing can increas membrane fouling due PAC being a potential
GAC	• Improve methane production • Enhance COD, SMP and pharmaceuticals removal • GAC fluidization has lower energy requirement than gas sparging • Scouring effect • Recover can be done by thermal process after adsorption capacity exhausted	• The abrasion can aggra fouling • Large particles require energy for fluidization
Biochar	• Improve hydrogen and methane production by enrichment of microorganisms • Decrease SMP and proteins of EPS • Enhance COD removal efficiency and sludge granulation	• Less efficient specific s area than AC due to no activation
Waste yeast	• High degradation capacity of phenolic and toxic compounds • Low tendency to adhere on membrane surface • Enhance biogas production	• No significant change i accumulation • Increase SMP and arom group with high molecu weight
Iron	• Enhance sludge granulation and stabilization • Improve methane production and COD removal • Eliminate H_2S gas • Enhance mixed liquor filterability • Advantageous for long-term operation due to the formation of a precipitate	• Formation of thick cake • Cause more severe me fouling due to the high of Fe-rich fouling layer
Calcium	• Enhance bioflocculation • Improve accumulation of biomass and degradation of butyrate and acetate acid • Reduce fine particles, SMP,	• Overdosing can lead to precipitation, declined methanogenic activity inorganic fouling

	EPS and colloids	
Polyaluminum chloride	• High charge neutralization capacity can lead to enlarged floc size and high filtration performance • Improve the abundance of anaerobic microorganisms and hydrogen yield • Reduce the composition rate of carbohydrate in SMP and EPS • The cake layer become more porous and looser	• High concentration can inhibit SCFAs production and decrease phosphorous
Zeolite	• High ion exchange capacity can enhance ammonia and heavy metals removal • Improve methane production, COD, nitrogen and phosphorous removal • Enhance sludge settlement by bacterial attachment and aggregation	
Beads	• Fluidization of PET beads can act as turbulence promotors and scouring media • Polymer-based gel beads have cost effectiveness, high bio-compatibility, high stability for long-term use, and porous structure for microbial attachment and aggregation • The size and density can be controlled by different synthesis conditions • Increase COD removal rate and methane production	• Fluidization of glass beads can damage membrane by abrasion

3. Future perspectives

Membrane fouling is one of the most challenging issues in operating MBR processes. Pretreatment of feed wastewater can effectively mitigate membrane fouling by changing the feed properties. The addition of fouling reduction enhancers to bioreactors as adsorbents, coagulants/flocculants and suspended carriers can significantly modify the feed characteristics. To date, there have been investigations of many different enhancers applied to aerobic MBRs for the purpose of fouling control and improvement of bioreactor performance. However, only a limited number of studies were available to investigate the effects of enhancers' addition in AnMBRs and only a few types of enhancers were studied previously. Hence, it is necessary and important not only to study the application of novel enhancers in anaerobic treatment, but also to understand the fouling reduction mechanisms of each enhancer under anaerobic condition.

As discussed in this review, previous studies have applied several enhancers to AnMBRs. The addition of activated carbon, such as PAC and GAC, has proven to be effective solution to alleviate membrane fouling. Both PAC and GAC could act as a supporting medium for the growth of anaerobic microorganisms due to its high adsorption capacity. PAC could effectively adsorb colloids and dissolved organic matters in AnMBRs, while GAC mainly adopted as fluidized media and could scour membrane as well as enhance methane production. Throughout the organic and inorganic enhancers, biochar and zeolite could be applied as adsorbents, while calcium was able to act as flocculants. In addition, iron and PACl were added as coagulants, and waste yeast and beads could perform as co-substrates and biocarriers, respectively.

However, overdosing or large particle size of enhancers caused more severe membrane fouling and deterioration of removal performances, due to their potential to become a foulant. Although optimal PAC replenishment ratio for effective fouling mitigation in aerobic MBR was reported to be 1.67%, the refreshment ratio of PAC or GAC in AnMBR remains as a challenging issue [155]. Thus, further studies regarding the optimization of the dosage as well as replacement ratio of enhancers should be carried out for the best improvement of performance as well as controlling membrane fouling. Furthermore, more studies on the different types of novel enhancers, such as waste yeast and beads, are necessary, because there are much less number of studies compared to that of activated carbon. Additionally, more research regarding the long-term effect of enhancers is also required, since they may influence negatively on AnMBR performance during long-term operation. Moreover, it is also important to consider the lifespan of membrane itself, when enhancer is added. The addition of particulate enhancers, such as PAC, GAC and biochar, could greatly mitigate irreversible membrane fouling and prolong the lifespan of membrane. However, comparatively large particle size or fluidization of GAC and glass beads could lead to abrasion and damage on membrane. Therefore, more research on the membrane lifespan with enhancers is also required for better understanding on the performance of enhancers.

In spite of many applications and research have been carried out in AnMBR with various enhancers, most of the research is confined to lab scale experiments. The major obstacle limiting scale up and wider application of AnMBRs can be membrane fouling, as membrane is one of the main contributors to capital and operational costs in AnMBRs. The capital cost of a full-scale submerged AnMBR system was about 800 USD/m^3/day. It was also estimated that almost 72.3% of capital cost accounted for membrane fraction with the assumed capacity of 20000 m^3/day, which was higher than that of aerobic MBR system (25-60%) [1, 156]. On the other hand, the operational cost including gas scouring, pumping and sludge disposal for submerged AnMBR was reported to be almost one third of that of aerobic MBR system, which was about USD 235000/year and USD 822741/year, respectively [156]. Thus, the full-scale AnMBR system could be economically feasible by adopting solutions such as low cost filters.

Since the energy requirement for membrane fouling control in AnMBR accounted more than 75% of the total energy requirement, it is important to apply efficient fouling control strategies in terms of energy consumption and costs. As discussed in section 2.1.2, GAC fluidization had lower energy consumption than gas sparging. The addition of enhancers such as $FeCl_3$ required much lower energy of 0.08 kWh/m^3 due to the lack of rotation [157]. In terms of cost, activated carbon (0.6-20 USD/kg) and biochar (0.2-0.5 USD/kg) could be relatively cheaper compared to other chemical enhancers [158]. Therefore, careful consideration and further analysis of enhancers for practical AnMBR application should be conducted.

To sum up, further studies on the optimisation of the adequate dosage and the impact of different particle size of each enhancer should be done in the near future, accompanied by the investigation of long-term impact. Moreover, the interaction mechanisms between the enhancers and anaerobic microbial activities also needs further exploration to better understand the influences on membrane fouling mitigation. Since the scale-up from bench-scale experiment to full-scale application is not simple, it is necessary to research further for the wide implementation of full-scale AnMBR with minimized membrane fouling.

3. Conclusion

The main conclusions in this review are as follows:

- The addition of fouling reduction enhancers, including activated carbon, biochar, zeolite, and polyaluminum chloride, could effectively alleviate membrane fouling in AnMBRs.
- Enlarged floc size and decreased soluble organics mainly contributed to the mitigation of fouling.
- Overdosing or large particle size of enhancers could lead to contrary result due to their potential to be a foulant.

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